DECLARATION

I, Chizuko IKEDA, declare that I reside at 5-50-301, Makita-cho, Takatsuki-shi, OSAKA 569-0855 JAPAN;

That I am familiar with the English and Japanese languages;

That I have prepared a translation of Japanese Patent Application No. 371173/1999, "難燃性樹脂組成物, FLAME-RETARDANT RESIN COMPOSITION"; said translation thereof being attached hereto and made a part of this declaration;

That to the best of my knowledge and belief, the attached translation is accurate and fairly reflects the contents and meaning of the foregoing Japanese language document.

I declare, under penalty of perjury under the laws of the United States of America, that the foregoing is true and correct.

Executed, on August 26, 2003.

Chizuko Ikeda

Chizuko IKEDA

PATENT OFFICE

JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy of the following application as filed with this Office.

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	Otsuka Chemical Co., Ltd.

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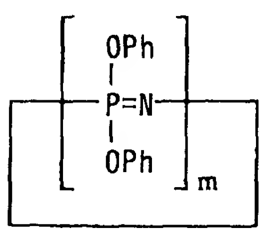
[Title of the Invention] FLAME-RETARDANT RESIN COMPOSITION

[Claims]

[Claim 1] A flame-retardant resin composition which comprises a flame retardant comprising a phosphazene compound and a phenolic resin, and a polyalkylene terephthalate resin, wherein the phosphazene compound comprises at least one member selected from the group consisting of:

(1) a cyclic phenoxyphosphazene compound of the formula :

[Formula 1]

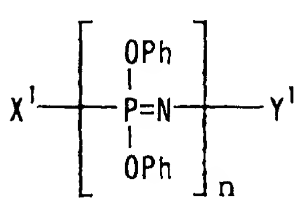


(1)

wherein m is an integer of 3 to 25, and Ph denotes a phenyl group,

(2) a linear phenoxyphosphazene compound of the formula :

[Formula 2]



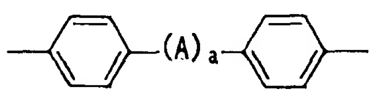
(2)

wherein X^1 represents the group $-N=P(OPh)_3$ or the

group -N=P(O)OPh; Y^1 represents the group $-P(OPh)_4$ or the group $-P(O)(OPh)_2$: n is an integer of 3 to 10,000; and Ph has the same meaning as defined in the formula (1), and

(3) a crosslinked phenoxyphosphazene compound which is formed by crosslinking at least one phenoxyphosphazene compound selected from the group consisting of the cyclic phenoxyphosphazene compound (1) and the linear phenoxyphosphazene compound (2) with at least one crosslinking group selected from the group consisting of o-phenylene group, m-phenylene group, p-phenylene group, and a bisphenylene group shown by the general formula (3):

[Formula 3]



(3)

wherein A represents $-C(CH_3)_2-$, $-SO_2-$, -S- or -O-, and a denotes 0 or 1, and

wherein the proportion of the crosslinking group in the crosslinked phenoxyphosphazene compound is 50 to 99.9 mol% on the basis of the total phenyl groups in the phenoxyphosphazene compounds (1) and (2), and the crosslinked phenoxyphosphazene compound does not contain a free hydroxyl group in its molecule.

[Claim 2] A composition according to Claim 1, wherein the polyalkylene terephthalate resin comprises a polyethylene terephthalate or a polybutylene terephthalate.

[Claim 3] A composition according to Claim 1,

wherein the flame retardant containing the phosphazene compound and the phenolic resin in a ratio (the phosphazene compound/the phenolic resin) of 20/80 to 80/20 (weight ratio) is used at 5 to 80 parts by weight relative to 100 parts by weight of the polyalkylene terephthalate resin.

[Claim 4] A composition according to Claim 1, which further comprises a nitrogen-containing flame retardant and/or a carbonizable resin.

[Claim 5] A composition according to Claim 1, which further comprises at least one member selected from the group consisting of an antioxidant, a drip inhibitor and a filler.

[Detailed Description of the Invention]

[0001]

[Technical Field of the Invention]

The present invention relates to a flame-retardant resin composition comprising a polyalkylene terephthalate resin.

[0002]

[Background Art]

A polyalkylene terephthalate resin such as a polybutylene terephthalate has excellent mechanical and electrical properties, weather resistance, water resistance, and resistance to chemicals and solvents. Therefore, the resin is used as an engineering plastic in a variety of applications such as electric or electronic device parts, mechanical device parts and automotive parts.

While, the resin is required to be flame-retardant from viewpoint of safety as the field of their uses expands. In general, there is known a method for rendering the resin flame-retardant by adding a flame retardant composed of a halogen-containing compound and/or an antimony-containing compound to the resin.

[0003]

Japanese Patent Application Laid-Open No. 168297/1998 (JP-10-168297A) discloses a flame-retardant resin composition comprising a thermoplastic polyester resin, a polycarbonate-series resin, an organic phosphorus-series flame retardant which is a phosphate-series flame retardant. Japanese Patent Application Laid-Open No. 195283/1998 (JP-10-195283A) discloses a polyester resin composition which is rendered flame-retardant by combining a phosphate having the specific structure, a novolak-type phenol resin and the specific oxide of a metal such as iron, cobalt, nickel or copper in suitable amounts. However, the phosphate-series flame retardant does not contain a harmful halogen, but a large amount of the flame retardant is required because of their inferior flame retardancy to the halogen-containing flame retardant. Thus, a bleed out and deterioration of the mechanical properties of the resin are caused and the mechanical properties cannot be improved accompanied with the flame retardancy.

[0004]

Moreover, Japanese Patent Application Laid-Open No. 181268/1999 (JP-11-181268A) discloses that addition of 1.5 to 15 parts by weight of a phosphazene compound and 0.5 to 30 parts by weight of talc and/or mica relative to 100 parts by weight of a resin mixture containing an aromatic polycarbonate-series resin and a thermoplastic polyester-series resin in the proportion of 90/10 to 50/50 (weight ratio) renders the resin mixture flame-retardant. However, the resin composition composed of the aromatic polycarbonate has a problem of resistance to solvents, and further, the moldability is lowered because of the inferior melt-flowability upon molding.

[0005]

Incidentally, Japanese Patent Application Laid-Open No. 181429/1999 (JP-11-181429) discloses that either a thermoplastic resin (e.g., polyethylene terephthalate, polybutylene terephthalate, polycarbonate) or a thermosetting resin (e.g., phenol resin) is rendered flame-retardant with use of the specific phosphazene compound (e.g., a cyclic phosphazene compound, a linear phosphazene compound, a crosslinked phosphazene compound obtained by crosslinking the cyclic and/or the linear phosphazene compound with the specific group) as a flame retardant. However, in the case of rendering a polyethylene terephthalate or a polybutylene terephthalate flame-retardant, the phosphazene compound alone cannot sufficiently impart the flame-retardancy to

the resin.

[0006]

[Problems to be Solved by the Invention]

Accordingly, it is object of the present invention to provide a resin composition having a high flame-retardancy without deteriorating the properties of a polyalkylene terephthalate resin.

[0007]

[Means to Solve the Problems]

The inventors of the present invention did intensive research, and finally found that a polyalkylene terephthalate resin can be rendered highly flame-retardant without deteriorating mechanical properties with use of a flame retardant comprising a phenoxyphosphazene compound and a phenolic resin in combination. The present invention was accomplished based on the above findings.

[0008]

That is, the flame-retardant resin composition comprises a flame retardant comprising a phosphazene compound and a phenolic resin, and a polyalkylene terephthalate resin (e.g., a polyethylene terephthalate, a polybutylene terephthalate). The phosphazene compound comprises (1) a cyclic phenoxyphosphazene compound, (2) a linear phenoxyphosphazene compound, or (3) a crosslinked phenoxyphosphazene compound which are shown below.

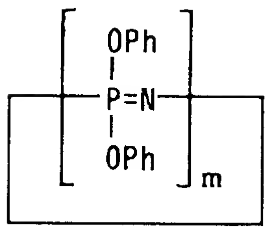
[0009]

(1) a cyclic phenoxyphosphazene compound of the

formula :

[0010]

[Formula 4]



(1)

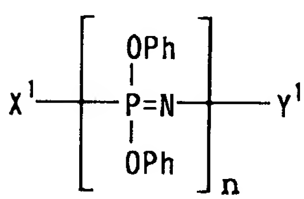
[0011]

wherein m is an integer of 3 to 25, and Ph denotes a phenyl group,

(2) a linear phenoxyphosphazene compound of the formula :

[0012]

[Formula 5]



(2)

[0013]

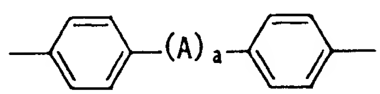
wherein X^1 represents the group $-N=P(OPh)_3$ or the group $-N=P(O)OPh$; Y^1 represents the group $-P(OPh)_4$ or the group $-P(O)(OPh)_2$; n is an integer of 3 to 10,000; and Ph has the same meaning as defined in the formula (1), and

(3) a crosslinked phenoxyphosphazene compound which is formed by crosslinking at least one phenoxyphosphazene compound selected from the group consisting of the cyclic phenoxyphosphazene compound (1) and the linear

phenoxyphosphazene compound (2) with at least one crosslinking group selected from the group consisting of o-phenylene group, m-phenylene group, p-phenylene group, and a bisphenylene group shown by the general formula (3):

[0014]

[Formula 6]



(3)

[0015]

wherein A represents $-C(CH_3)_2-$, $-SO_2-$, -S- or -O-, and a denotes 0 or 1, and

wherein the proportion of the crosslinking group in the crosslinked phenoxyphosphazene compound is 50 to 99.9 mol% on the basis of the total phenyl groups in the phenoxyphosphazene compounds (1) and (2), and the crosslinked phenoxyphosphazene compound does not contain a free hydroxyl group in its molecule.

The proportion of the phosphazene compound relative to the phenolic resin (the phosphazene compound/the phenolic resin) in the flame retardant may be 20/80 to 80/20 (weight ratio), and the flame retardant may be used at 5 to 80 parts by weight relative to 100 parts by weight of the polyalkylene terephthalate resin. The resin composition may further comprise a nitrogen-containing flame retardant, a carbonizable resin, an antioxidant, a drip inhibitor, a filler, and others.

[0016]

[Preferred Embodiment of the Invention]

[Polyalkylene terephthalate resin]

The polyalkylene terephthalate resin includes a homopolyester or a copolyester containing an alkylene terephthalate as a main component (e.g., about 50 to 100% by weight, preferably about 75 to 100% by weight). The homopolyester includes, for example, poly1,4-cyclohexane dimethylene terephthalate (PCT), polyethylene terephthalate (PET), polypropylene terephthalate (PPT), polybutylene terephthalate (PBT). A copolymerizable monomer of the copolyester includes, for example, an alcohol component such as (poly)ethylene glycol and (poly)propylene glycol, a carboxylic acid component such as adipic acid and isophthalic acid. These polyalkylene terephthalates can be used singly or in combination. The preferred polyalkylene terephthalate resin is a polyC_{2-4}alkylene terephthalate such as polyethylene terephthalate and polybutylene terephthalate.

[0017]

The number average molecular weight of the polyalkylene terephthalate resin is not particularly limited, and can be selected within the range of, for example, about 5×10^3 to 100×10^4, preferably about 1×10^4 to 70×10^4, and more preferably about 1.2×10^4 to 30×10^4.

[0018]

The polyalkylene terephthalate resin can be prepared by a conventional method, for example, a transesterification method or a direct esterification method with use of an alkylene glycol and terephthalic acid.

[0019]

[Flame retardant]

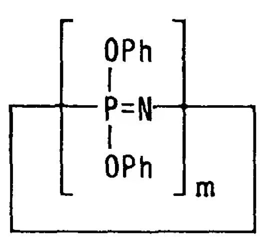
The flame retardant of the present invention comprises a phosphazene compound (a cyclic phenoxyphosphazene compound, a linear (or chain) phenoxyphosphazene compound, a crosslinked phenoxyphosphazene compound) and a phenolic resin. The flame-retardant comprises the phosphazene compound and the phenolic resin so that the high flame-retardancy can be imparted to the polyalkylene terephthalate resin without deteriorating mechanical properties.

[0020]

The cyclic phenoxyphosphazene compound includes a compound shown by the following general formula (1):

[0021]

[Formula 7]



(1)

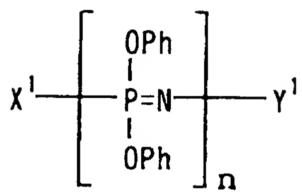
[0022]

wherein m is an integer of 3 to 25, and Ph denotes a phenyl group.

The linear phenoxyphosphazene compound includes a compound shown by the general formula (2):

[0023]

[Formula 8]



(2)

[0024]

wherein X^1 represents the group $-N=P(OPh)_3$ or the group $-N=P(O)OPh$; Y^1 represents the group $-P(OPh)_4$ or the group $P(O)(OPh)_2$; n is an integer of 3 to 10,000; and Ph has the same meaning as defined in the formula (1).

The crosslinked phenoxyphosphazene compound includes a compound which is formed by crosslinking at least one phenoxyphosphazene compound selected from the group consisting of the cyclic phenoxyphosphazene compound (1) and the linear phenoxyphosphazene compound (2) with a divalent crosslinking group. Incidentally, when a pair of phenoxyphosphazene compounds is crosslinked with the crosslinking group, the divalent crosslinking group is introduced in lieu of a pair of Ph groups.

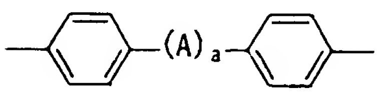
[0025]

The divalent crosslinking group includes a phenylene group (o-phenylene group, m-phenylene group, p-phenylene group), and a bisphenylene group shown by the following general formula (3). Incidentally, these crosslinking

groups can be used singly or in combination.

[0026]

[Formula 9]

 (3)

[0027]

wherein A represents $-C(CH_3)_2-$, $-SO_2-$, $-S-$ or $-O-$, and a is 0 or 1.

The proportion of the crosslinking group is about 50 to 99.9 mol% on the basis of the total phenyl groups in the phenoxyphosphazene compounds (1) and (2).

[0028]

Incidentally, the crosslinked phenoxyphosphazene compound does not substantially contain a free hydroxyl group in its molecule.

[0029]

These phosphazene compounds can be used singly or in combination.

[0030]

The cyclic and linear phenoxyphosphazene compounds shown by the general formulae (1) and (2) can be synthesized by the method described in "Phosphorus-Nitrogen Compounds" by H.R.Allcock, published by Academic Press, (1972), "Inorganic Polymers" by J.E.Mark, H.R.Allcock, R.West, published by Prentice-Hall International, Inc.,(1992).

[0031]

For example, a mixture of a compound of the general

formula (1) wherein the group OPh is substituted by a chlorine atom (Cl) and m is an integer of 3 to 25 (a cyclic dichlorophosphazene oligomer), and a compound of the general formula (2) wherein the group OPh is substituted by a chlorine atom and n is an integer of 3 to 25 (a chain dichlorophosphazene oligomer) can be obtained by a reaction of phosphorus chloride (e.g., phosphorus trichloride, phosphorus pentachloride) and ammonium chloride, and if necessary a chlorine (in particular, in the case of using phosphorus trichloride as the phosphorus chloride) in a chlorine-series solvent (e.g., chlorobenzene, tetrachloroethane). The cyclic and linear phenoxyphosphazene compounds represented by the general formulae (1) and (2) can be obtained by substituting a chlorine atom of the dichlorophosphazene oligomer mixture by phenol with use of an alkali metal phenolate (e.g., sodium phenolate).

[0032]

The reaction temperature in a reaction of phosphorus chloride and ammonium chloride is, for example, about 120 to 130°C.

[0033]

If necessary, the mixture of the dichlorophosphazene oligomer may be subjected to purification (e.g., distillation, recrystallization) or polymerization (ring-opening-polymerization of a cyclic dichlorophosphazene oligomer). By purifying the mixture

of the dichlorophosphazene oligomer, a single or sole compound of the cyclic dichlorophosphazene (e.g., hexachlorocyclotriphosphazene, octachlorocyclotetraphosphazene, decachlorocyclopentaphosphazene) can be obtained. Therefore, by substituting the single compound with a phenol, the cyclic phenoxyphosphazene compound such as hexaphenoxycyclotriphosphazene, octaphenoxycyclotetraphosphazene, and decaphenoxycyclopentaphosphazene can be obtained.

[0034]

While, a cyclic dichlorophosphazene oligomer is ring-opening-polymerized to obtain a compound of the general formula (2) wherein the group OPh is substituted with a chlorine atom and n is an integer of 3 to 10,000. Therefore, by substituting the compound with a phenol, the linear phenoxyphosphazene compound shown by the general formula (2) can be obtained.

[0035]

The ring-opening-polymerization of the cyclic dichlorophosphazene oligomer can be carried out, for example, by heating to 220 to 250°C.

[0036]

The crosslinked phenoxyphosphazene compound can be produced by substituting (crosslinking) a chlorine atom of the dichlorophosphazene oligomer with an aromatic dihydroxy compound in lieu of by substituting the chlorine

atom with a phenol in the production process of the cyclic phosphazene compound (1) or the linear phosphazene compound (2).

[0037]

In more detail, a partial-substituted compound in which one part of chlorine atoms of the dichlorophosphazene compound is substituted with a phenol and one part thereof is substituted with an aromatic dihydroxy compound, and one part thereof is retained as chlorine atom, is obtained by reacting the dichlorophosphazene compound (e.g., cyclic dichlorophosphazene oligomer, linear dichlorophosphazene oligomer), an alkali metal salt of a phenol and an alkali metal salt of an aromatic dihydroxy compound (the first stage reaction). Then, the partial-substituted compound is reacted with an alkali metal salt of phenol (the second stage reaction) so that the crosslinked phenoxyphosphazene compound can be obtained. Thus the resulting crosslinked phenoxyphosphazene compound seldom contains a free hydroxyl group since all of hydroxyl groups of the aromatic dihydroxy compound are reacted with dichlorophosphazene compounds.

[0038]

As the aromatic dihydroxy compound, use can be made of a compound having one or not less than two benzene rings in its molecule and two hydroxyl groups, more concretely, a compound having the above crosslinking group (o-phenylene group, m-phenylene group, p-phenylene group, a

group shown by the general formula (3)). The preferred aromatic dihydroxy compound includes resorcinol, hydroquinone, catechol, 4,4'-isopropylidenediphenol (bisphenol-A), 4,4'-sulfonyldiphenol (bisphenol-S), 4,4'-thiodiphenol, 4,4'-oxydiphenol, 4,4'-diphenol and the like. The aromatic dihydroxy compound can be used singly or in combination.

[0039]

The alkali metal constituting the alkali metal salt includes sodium, potassium, lithium and the like, preferably sodium and lithium.

[0040]

In the first stage reaction, the total amount of an alkali metal salt of phenol and an alkali metal salt of an aromatic dihydroxy compound is usually about 0.05 to 0.9 equivalent, and preferably about 0.1 to 0.8 equivalent based on the chlorine content of the dichlorophosphazene oligomer. When the amount of the alkali metal salt is significantly less than 0.05 equivalent, the degree of crosslinking is insufficient. While, when the amount of the alkali metal salt is dramatically more than 0.9 equivalent, a free hydroxyl group (a hydroxyl group at one side of the dihydroxy compound) is introduced into the crosslinked phenoxyphosphazene compound.

[0041]

The ratio of the alkali metal salt of the aromatic dihydroxy compound relative to that of phenol is not

particularly limited, can be suitably selected within a wide range and is usually the former / the latter = about 1/2000 to 1/4 (molar ratio). When the ratio is remarkably less than 1/2000, the degree of crosslinking is insufficient. While, when the ratio is dramatically more than 1/4, the crosslink proceeds too much, so that the solubility and meltability of the crosslinked phenoxyphosphazene compound are deteriorated and the dispersability in a resin is inadequate.

[0042]

The first stage reaction may be carried out in a solvent (e.g., an aromatic hydrocarbon such as toluene, a halogenated aromatic hydrocarbon such as chlorobenzene).

[0043]

The reaction temperature in the first stage reaction is usually from a room temperature to about 150°C.

[0044]

In the second stage reaction, the amount of the alkali metal salt of phenol is usually about 1 to 1.5 equivalents, preferably about 1 to 1.2 equivalents based on the chlorine content of dichlorophosphazene oligomer.

[0045]

The proportion of the phosphazene compound is, for example, about 1 to 40 parts by weight, preferably about 1 to 30 parts by weight, and more preferably about 5 to 25 parts by weight relative to 100 parts by weight of the polyalkylene terephthalate resin.

[0046]

As the phenolic resin, a variety of resins having a phenol residue as a constituting unit can be used and include for example, novolak resins. The novolak resin includes a compound obtained by a reaction of a phenol (e.g., phenol, a phenol substituted with a C_{1-10}alkyl group such as cresol, ethylphenol, propylphenol, butylphenol, or octylphenol; cyanophenol) and an aldehyde (e.g., formaldehyde, acetaldehyde, propionaldehyde, in particular, formaldehyde).

[0047]

The condensation reaction of the phenol and the aldehyde is usually carried out in the presence of an acid catalyst such as an inorganic acid (e.g., hydrochloric acid, sulfuric acid) and an organic acid (e.g., p-toluenesulfonic acid, oxalic acid). The ratio of the phenol relative to the aldehyde is the former/the latter = about 1/0.6 to 1/1 (molar ratio).

[0048]

The phenolic resin can be used singly or in combination. The preferred phenolic resin is a phenol-novolak resin.

[0049]

Incidentally, a part or all of the phenolic hydroxyl groups of the phenolic resin may be optionally modified with a phosphorus compound such as a phosphoric acid or a phosphate.

[0050]

The number average molecular weight of the phenolic resin is not particularly limited and can for example be selected within the range of about 300 to 5 x 10^4, preferably about 300 to 1 x 10^4, and more preferably about 300 to 8,000.

[0051]

The ratio of the phenolic resin used in the flame retardants is, for example, about 1 to 40 parts by weight, preferably about 1 to 30 parts by weight, and more preferably about 3 to 25 parts by weight (in particular, about 5 to 20 parts by weight) relative to 100 parts by weight of the polyalkylene terephthalate resin.

[0052]

Moreover, the ratio of the phosphazene compound relative to the phenolic resin in the flame retardant is, for example, the former/the latter = about 5/95 to 95/5 (weight ratio), preferably about 20/80 to 80/20 (weight ratio), more preferably about 30/70 to 70/30 (weight ratio), and may be about 60/40 to 80/20 (weight ratio) and preferably about 50/50 to 75/25 (weight ratio).

[0053]

Since the flame retardant of the present invention contains a phenolic resin, the flame-retardancy can be imparted to the polyalkylene terephthalate resin with inhibiting the decline in a molecular weight and a mechanical property (e.g., strength, impact resistance) of the polyalkylene terephthalate resin. In particular,

the phosphazene compound is combined with the phenolic resin so that the polyalkylene terephthalate resin is provided with higher flame-retardant compared to the case of using the phosphazene compound singly. Moreover, since the flame retardant does not contain a halogen, there is no possibility that a hydrogen halide which is poisonous gas generates upon decomposition or burning, and that corrosion of a mould and deterioration of a resin occur upon molding the resin.

[0054]

The proportion of the flame retardant in the resin composition is not particularly limited as far as the property of the polyalkylene terephthalate resin is not deteriorated and is about 1 to 100 parts by weight, preferably about 5 to 80 parts by weight, more preferably about 5 to 60 parts by weight, and particularly about 10 to 50 parts by weight relative to 100 parts by weight of the polyalkylene terephthalate resin. When the amount of the flame retardant is less than 1 part by weight, it is difficult that the flame-retardancy is imparted. When the amount of the flame retardant is more than 100 parts by weight, the mechanical strength and moldability of a molded article obtained from the resin composition are deteriorated.

[0055]

The flame retardant containing the phosphazene compound and the phenolic resin in a ratio (the former/the

latter) of about 20/80 to 80/20 (weight ratio) is usually employed at 5 to 80 parts by weight relative to 100 parts by weight of the polyalkylene terephthalate resin.

[0056]

The polyalkylene terephthalate resin composition of the present invention may optionally contain an additive (for example, other flame retardant, a carbonizable resin, a drip inhibitor, an antioxidant). The content of the additive is, for example, about 0.01 to 20 parts by weight, and preferably about 0.1 to 10 parts by weight relative to 100 parts by weight of the polyalkylene terephthalate resin. Moreover, the content of the carbonizable resin may be about 0 to 100 parts by weight, preferably about 1 to 80 parts by weight, and more preferably about 10 to 60 parts by weight relative to 100 parts by weight of the polyalkylene terephthalate resin.

[0057]

Among the additives, as the other flame retardants, there may be mentioned a nitrogen-containing flame retardant (e.g., melamine, guanamine, melamine cyanurate, guanamine cyanurate), an organic phosphorus-series flame retardant [for example, a phosphate (e.g., triphenyl phosphate, tricresyl phosphate); a polyphosphate (hydroquinone bis(diphenyl phosphate), resorcinol bis(dixylyl phosphate), bisphenol-A bis(dixylyl phosphate)], an inorganic phosphorus-series flame retardant (e.g., a red-phosphorus which may be coated with

a resin, phosphoric acid salt), a sulfur-containing flame retardant, a silicon-containing flame retardant (e.g., (poly)organosiloxane), an inorganic flame retardant (e.g., metal oxide, metal hydroxide), or others.

[0058]

The carbonizable resin includes a resin having an aromatic ring. As such the aromatic ring-containing resin, there may be exemplified a polycarbonate, a polyarylate, a phenoxy resin, a polyphenylene oxide, a polyphenylene sulfide, or others. These carbonizable resins can be used singly or in combination.

[0059]

The drip inhibitor includes a fluororesin such as a homo- or copolymer of a fluorine-containing monomer, or a copolymer of the fluorine-containing monomer with the other copolymerizable monomer. As the fluororesin, there may be mentioned polytetrafluoroethylene, polychlorotrifluoroethylene, polyvinylidene fluoride, tetrafluoroethylene-hexafluoropropylene copolymer, tetrafluoroethylene-perfluoroalkylvinylether copolymer, ethylene-tetrafluoroethylene copolymer, ethylene-chlorotrifluoroethylene copolymer and the like.

[0060]

As the antioxidant, there may be mentioned, for example, hindered phenol-series antioxidants (e.g., 2,6-di-t-butyl-p-cresol, 2,2'-methylenebis(4-methyl-6-t-butylphenol), 2,2'-thiobis(4-methyl-6-t-butylphenol),

4,4'-thiobis(6-t-butyl-m-cresol), and pentaerythritol-tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate]), amine-series antioxidants (e.g., naphthylamine), phosphorus-series antioxidants (e.g., a phosphite such as bis(2,4-di-t-butylphenyl) pentaerythritol diphosphite, and bis(2,6-di-t-butyl-4-methylphenyl) pentaerythritol diphosphite, a phosphonite such as tetrakis(2,4-di-t-butylphenyl)-4,4'-biphenylenediphosphonite).

[0061]

Moreover, the resin composition of the present invention may optionally contain the other additives such as a lubricant, a mold-releasing agent, a plasticizer, a flame-retardant auxiliary, an ultraviolet ray absorbing agent, a pigment, a dye, an antistatic agent, a dispersing agent, a compatibilizer, an antibacterial agent and the like.

[0062]

Further, the resin composition may optionally contain a filler (e.g., kaolin, mica, talc, calcium carbonate, titanium oxide, glass flake, various metal foils, glass fiber, carbon fiber). The amount of the filler is about 5 to 100 parts by weight, and preferably about 10 to 80 parts by weight relative to 100 parts by weight of the polyalkylene terephthalate resin.

[0063]

The resin composition of the present invention may

be whichever a powdered mixture or a melt mixture, and can be prepared by mixing the polyalkylene terephthalate resin, the flame retardant, and optionally, the additive by means of a conventional method.

[0064]

The resin composition of the present invention can be melt-kneaded and molded by a conventional method such as extrusion molding, injection molding, and compression molding. Since the molded article which is formed is superior in a flame-retardancy and mold-processability, the article can be utilized in a variety of applications. For example, the article is favorable for use in electric or electronic device parts, mechanical device parts and automotive parts.

[0065]

[Effects of the Invention]

According to the present invention, since the flame retardant comprising the specific phosphazene compound and the phenolic resin in combination is used, it is made possible to give high flame-retardancy to the polyalkylene terephthalate resin without using a halogen-containing flame retardant. In particular, according to the present invention, it is possible to make the polyalkylene terephthalate resin highly flame-retardant without adversely affecting properties of the flame retardant polyalkylene terephthalate resin.

[0066]

[Examples]

The following examples are intended to describe this invention in further detail and should by no means be interpreted as defining the scope of the invention.

[0067]

In Examples and Comparative Examples, a polyester resin, a flame retardant (a compound, a phenolic resin), and optionally an additive (a nitrogen-containing flame retardant, a carbonizable resin, an antioxidant, a drip inhibitor), and a filler, which are shown below were used, as a phosphazene compound.

[0068]

[Polyalkylene terephthalate A]

A-1 : polybutylene terephthalate [Duranex, intrinsic viscosity 1.0, manufactured by Polyplastics, Co. Ltd.]

A-2 : polybutylene terephthalate [Duranex, intrinsic viscosity 0.75, manufactured by Polyplastics, Co. Ltd.]

A-3 : polyethylene terephthalate (BelPet EFG10, manufactured by Kanebo, Ltd.)

[Phosphazene compound B]

B-1 to B-5 : phenoxyphosphazene compounds obtained by the following synthesis examples 1 to 5.

[Phenolic resin C]

C-1 : novolak resin [Tamanol 759, manufactured by Arakawa Chemical Industries, Ltd.]

C-2 : novolak resin [Sumilite Resin PR-53195, manufactured by Sumitomo Durez Co., Ltd.]

[Nitrogen-containing flame retardant D]

D-1 : melamine cyanurate [MC610, manufactured by Nissan Chemical Industries, Ltd.]

[Carbonizable resin E]

E-1 : polycarbonate [Panlite L1225, manufactured by Teijin Chemicals, Ltd.]

[Antioxidant F]

F-1 : pentaerythritol tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate][Irganox 1010, manufactured by Ciba-Geigy Ltd.]

F-2 : bis(2,6-di-t-butyl-4-methylphenyl)pentaerythritol diphosphite [Adekastab PEP36, manufactured by Asahi Denka Kogyo K.K.]

F-3 : tetrakis(2,4-di-t-butylphenyl)-4,4'-biphenylenediphosphonite [Sandostab P-EPQ, manufactured by Sandoz.]

[Drip inhibitor G]

G-1 : polytetrafluoroethylene

[Filler H]

H-1 : glass fiber (diameter: 10 μm, chopped strand of 3 mm length)

Synthesis Example 1 (synthesis of a cyclic phenoxyphosphazene compound (B-1))

Reference ; H.R.Allcock, "Phosphorus-Nitrogen Compounds", Academic Press, (1972)

To 580 g of chlorobenzene solution (20% by weight) containing 1.0 unit mol (115.9 g) of dichlorophosphazene

oligomer (a mixture of 62% by weight of a trimer and 38% by weight of a tetramer) was added a toluene solution containing sodium phenolate with stirring, and reacted for 4 hours at 110°C to obtain a cyclic phenoxyphosphazene compound. The amount of chlorine formed by hydrolysis after purification was 0.08% by weight.

[0069]

Synthesis Example 2 (Synthesis of a phenoxyphosphazene compound (B-2))

To a four neck flask (1L) equipped with a stirrer, a thermometer, and a reflux condenser were added 1.3 mol (123.0 g) of phenol and 500 mL of tetrahydrofuran to dissolve homogeneously. Then, 27.6 g of metal sodium was added at 25°C or less, and after adding, stirred for 6 hours at 61 to 68°C to prepare a sodium phenolate solution.

[0070]

To a four neck flask (2L) charged with 290 g of a chlorobenzene solution (20% by weight) containing 0.5 unit mol (58 g) of dichlorophosphazene oligomer (a mixture of 59% by weight of trimer, 12% by weight of tetramer, 11% by weight of pentamer and hexamer, 3% by weight of heptamer and 15% by weight of oligomers having eight or more repeating units) was added dropwise the sodium phenolate solution with stirring at 25°C or less. After adding dropwise, the reaction was carried out with stirring for 15 hours at 71 to 73°C. After the completion of the reaction, the reaction mixture was condensed and redissolved in 500

mL of chlorobenzene, washed three times with 5% by weight of NaOH aqueous solution, washed once with 5% by weight of sulfuric acid, washed once with 5% by weight of sodium hydrogencarbonate aqueous solution, washed twice with water and condensed to dryness to obtain 108 g of a light yellow wax-like material.

[0071]

The product was analyzed by GPC, and as a result, the weight-average molecular weight (Mw) was 810 in terms of polystyrene. The product was analyzed by TG/DTA, and as a result, the melting temperature was 103°C, the beginning temperature of decomposition was 330°C, and the 5% by weight loss temperature was 347°C. Moreover, the amount of the residual chlorine (chlorine formed by hydrolysis : Hy-Cl) was 0.09% by weight, and by the elemental analysis of phosphorus and CHN, it was determined that the product was the following compound :

[0072]

$[N{=}P(-OPh)_2]_n$

Synthesis Example 3 (Synthesis of a phenoxyphosphazene compound having a crosslinking structure with m-phenylene (B-3))

The mixture of 1.1 mol (103.5 g) of phenol, 1.1 mol (44.0 g) of sodium hydroxide, 50 g of water and 500 mL of toluene was heated with refluxing and water was exclusively removed from the system to prepare a toluene solution containing sodium phenolate.

[0073]

Parallel to the reaction, the mixture of 0.15 mol of (16.5 g) of resorcinol, 1.0 mol (94.1 g) of phenol, 1.3 mol (31.1 g) of lithium hydroxide, 52 g of water and 600 mL of toluene was heated with refluxing in a four neck flask (2L) and water was exclusively removed from the system to prepare a toluene solution containing resorcinol and lithium salt of phenol. To this toluene solution were added dropwise 580 g of a chlorobenzene solution (20% by weight) containing 1.0 unit mol (115.9 g) of dichlorophosphazene oligomer (62% by weight of trimer, 12% by weight of tetramer, 11% by weight of pentamer and hexamer, 3% by weight of heptamer, 12% by weight of oligomers having eight or more repeating units) at 30°C or less with stirring, and a reaction was carried out for 3 hours at 110°C with stirring. To the reaction mixture was added the toluene solution containing sodium phenolate and the reaction was continued for 4 hours at 110°C.

[0074]

After the completion of the reaction, the reaction mixture was washed three times with 3% by weight of sodium hydroxide aqueous solution (1.0 L), then washed three times with 1.0 L of water, and the organic layer was concentrated under reduced pressure. The resulting product was heated and vacuum dried for 11 hours at 80°C and 400 Pa or less to obtain 209 g of a white powder.

[0075]

In the resulting crosslinked phenoxyphosphazene compound, the chlorine amount formed by hydrolysis was 0.08% by weight, the weight-average molecular weight (MW) was 1080 in terms of polystyrene (determined by GPC analysis), and it was determined by the phosphorus content and the value of CHN elemental analysis that the composition of the final product was $[N=(-O-m-Ph-O-)_{0.15}(-O-Ph)_{1.7}]$. TG/DTA analysis was carried out, and as a result, the melting point was unclear, the beginning temperature of decomposition was 304°C, and 5% by weight loss temperature was 311°C. Moreover, the amount of the residual hydroxyl group was determined by an acetylation method, and as a result, the amount was below detection limits (a hydroxy equivalent per 1 g of a sample : 1×10^{-6} equivalent/g or less).

[0076]

Synthesis Example 4 (Synthesis of a phenoxyphosphazene compound having a crosslinking structure with 2,2-bis(p-oxyphenyl)isopropylidene group (B-4))

To a four neck flask (1L) containing 0.7 mol (65.9 g) of phenol and 500 mL of toluene was added 0.65 gram atom (14.9 g) of a subdivided metal sodium with maintaining the inner liquid temperature at 25°C and with stirring. After adding, the mixture was stirred for 8 hours at a temperature of 77 to 113°C until the metal sodium disappeared completely.

[0077]

Parallel to the reaction, to a four neck flask (3L) charged with 0.25 mol (57.1 g) of bisphenol A, 1.1 mol (103.5 g) of phenol and 800 mL of tetrahydrofuran (THF) was added 1.6 gram atom (11.1 g) of a subdivided metal lithium with maintaining the inner liquid temperature at 25°C or less and with stirring. After adding, the mixture was stirred for 8 hours at a temperature of 61 to 68°C until the metal lithium disappeared completely. To the slurry solution was added dropwise 1.0 unit mol (115.9 g) of dichlorophosphazene oligomer (concentration : 37% by weight, chlorobenzene solution 313 g, composition : 75% by weight of trimer, 17% by weight of tetramer, 6% by weight of pentamer and hexamer, 1% by weight of heptamer, 1% by weight of oligomers having eight or more repeating units) over 1 hour with maintaining the inner liquid temperature at 20°C or less and with stirring, and the reaction was carried out for 2 hours at 80°C. Then, with stirring and with maintaining the inner liquid temperature at 20°C, the sodium phenolate solution prepared separately was added over 1 hour, and the reaction was carried out at 80°C for 5 hours.

[0078]

After completion of the reaction, the reaction mixture was concentrated to remove off THF, and 1L of toluene was further added. The toluene solution was washed three times with 1 L of NaOH aqueous solution (2% by weight), then washed three times with 1.0 L of water, and the organic

layer was concentrated under reduced pressure. The resulting product was heated and vacuum dried for 11 hours at 80°C and 400 Pa or less to obtain 229 g of white powder.

[0079]

In the resulting crosslinked phenoxyphosphazene compound, a chlorine amount formed by hydrolysis was 0.07% by weight, and it was determined by the phosphorus content and the value of CHN elemental analysis that the final product was $[N=P(-O-Ph-C(CH_3)_2-Ph-O-)_{0.25}(-O-Ph)_{1.50}]$. The weight-average molecular weight (MW) was 1130 in terms of polystyrene (determined by GPC analysis). TG/DTA analysis was carried out, and as a result, the melting point was unclear, the beginning temperature of decomposition was 308°C, and 5% by weight loss temperature was 313°C. Moreover, the amount of the residual hydroxyl group was determined by an acetylation method, and as a result, the amount was below detection limits (a hydroxy equivalent per 1 g of a sample : 1×10^{-6} equivalent/g or less).

[0080]

Synthesis Example 5 (Synthesis of a phenoxyphosphazene compound having a crosslink structure with 4,4'-sulfonyldiphenylene(bisphenol-S residue) (B-5))

To a four neck flask (1L) charged with 0.4 mol (37.6 g) of phenol and 500 mL of THF was added 0.4 gram atom (9.2 g) of a subdivided metal sodium with maintaining the inner liquid temperature at 25°C and with stirring. After adding, the mixture was stirred for 5 hours at a temperature of

65 to 72°C until the metal sodium disappeared completely.

[0081]

Parallel to the reaction, to a four neck flask (1L) charged with 500 mL of tetrahydrofuran (THF) solution containing 1.70 mol (160.0 g) of phenol and 0.05 mol (12.5 g) of bisphenol-S was added 1.8 gram atom (41.4 g) of a metal sodium at 25°C or less with stirring. After adding, the temperature was elevated to 61°C over 1 hour and the mixture was stirred for 6 hours at a temperature of 61 to 68°C to prepare a mixture solution of sodium phenolate. This solution was added dropwise to 580 g of chlorobenzene solution (20% by weight) containing 1.0 unit mol (115.9 g) of dichlorophosphazene oligomer (a mixture of 62% by weight of trimer, 12% by weight of tetramer, 11% by weight of pentamer and hexamer, 3% by weight of heptamer, 12% by weight of oligomers having eight or more repeating units) with stirring and with cooling to 25°C or less, and the reaction was carried out for 5 hours at 71 to 73°C with stirring. Then, the sodium phenolate mixture solution prepared separately was added dropwise, and the reaction was carried out at 71 to 73°C for 3 hours.

[0082]

After completion of the reaction, the reaction mixture was concentrated, redissolved in 500 mL of chlorobenzene, washed three times with 5% by weight of NaOH aqueous solution, washed once with 5% by weight of sulfuric acid, washed once with 5% by weight of sodium

hydrogencarbonate, washed three times with water, and concentrated to dryness to obtain 218 g of light yellow wax-like material.

[0083]

In the resulting crosslinked phenoxyphosphazene compound, a chlorine amount formed by hydrolysis was 0.01% by weight or less, and it was determined by the phosphorus content and the value of CHN elemental analysis that the product was roughly $[N=P(-O-Ph-SO_2-Ph-O-)_{0.05}(-O-Ph)_{1.90}]$. The weight-average molecular weight (MW) was 1080 in terms of polystyrene (determined by GPC analysis). TG/DTA analysis was carried out, and as a result, the melting point was 103°C, the beginning temperature of decomposition was 320°C, and 5% by weight loss temperature was 334°C. Moreover, the amount of the residual hydroxyl group was determined by an acetylation method, and as a result, the amount was below detection limits (a hydroxy equivalent per 1 g of a sample : 1×10^{-6} equivalent/g or less).

[0084]

Examples 1 to 12 and Comparative Examples 1 to 11

Into polyalkylene terephthalate A were mixed phosphazene compound B, phenolic resin C, nitrogen-containing flame retardant D, carbonizable resin E, antioxidant F, drip inhibitor G, filler H and so on in the proportion shown by the Tables 1 to 4, and the mixture was kneaded at 240°C (in the case of Examples 1 to 6, 8 to 9, and Comparative Examples 1 to 8 and 10) or 270°C (in the

case of Example 7 and Comparative Example 9 adding polycarbonate; Examples 10 to 12 and Comparative Example 11 adding polyethylene terephthalate) for 5 minutes with the use of LABOPLASTMILL (manufactured by Toyo Seiki Seisaku-sho, Ltd.) to obtain a composition. The polyalkylene terephthalate resin composition was injection molded with a small-size molding machine to prepare a molded article for flame retardancy test (77 mm x 9.5 mm x 3 mm). The flame retardancy of the molded article was evaluated according to UL 94.

[0085]

The results are shown by Tables 1 to 4.

[0086]

[Table 1]

Table 1

	Ex.1	Ex.2	Ex.3	Ex.4	Ex.5	Ex.6
polyalkylene terephthalate resin A (parts by weight)	A-1 100	A-1 100	A-1 100	A-1 100	A-1 100	A-1 100
phosphazene compound B (parts by weight)	B-1 20	B-2 15	B-3 15	B-4 15	B-5 20	B-5 15
flame-retarding resin C (parts by weight)	C-1 15	C-1 15	C-2 15	C-2 15	C-2 15	C-2 15
the other flame retardant D (parts by weight)	-	D-1 7	D-1 7	D-1 7	-	D-1 7
carbonizable resin E (parts by weight)	-	-	-	-	-	-
antioxidant F (parts by weight)	F-1 0.15	F-1 F-2 0.15 0.15	F-1 F-3 0.15 0.15	F-1 F-3 0.15 0.15	F-1 F-2 0.15 0.15	F-1 F-3 0.15 0.15
drip inhibitor G (parts by weight)	G-1 0.7	G-1 0.7	G-1 0.7	G-1 0.7	G-1 0.7	G-1 0.7
filler H (parts by weight)	-	-	-	-	-	-
flame retardancy UL94	V-1	V-0	V-0	V-0	V-1	V-0

[0087]

[Table 2]

Table 2

	Ex.7	Ex.8	Ex.9	Ex.10	Ex.11	Ex.12
polyalkylene terephthalate resin A (parts by weight)	A-1 100	A-2 100	A-2 100	A-3 100	A-1 A-3 70 30	A-2 A-3 70 30
phosphazene compound B (parts by weight)	B-5 20	B-3 20	B-5 20	B-3 15	B-5 15	B-5 20
flame-retarding resin C (parts by weight)	C-2 10	C-2 20	C-2 20	C-2 15	C-2 15	C-2 20
the other flame retardant D (parts by weight)	D-1 10	D-1 10	D-1 10	D-1 7	D-1 7	D-1 10
carbonizable resin E (parts by weight)	E-1 30	-	-	-	-	-
antioxidant F (parts by weight)	F-1 F-3 0.2 0.2	F-1 F-3 0.2 0.2	F-1 F-3 0.2 0.2	F-1 F-3 0.2 0.2	F-1 F-3 0.15 0.15	F-1 F-3 0.2 0.2
drip inhibitor G (parts by weight)	G-1 1.0	G-1 1.0	G-1 1.0	G-1 1.0	G-1 1.0	G-1 1.0
filler H (parts by weight)	-	H-1 65	H-1 65	-	-	H-1 65
flame retardancy UL94	V-0	V-0	V-0	V-0	V-0	V-0

[0088]

[Table 3]

Table 3

	Comp.Ex. 1	Comp.Ex. 2	Comp.Ex. 3	Comp.Ex. 4	Comp.Ex. 5	Comp.Ex. 6
polyalkylene terephthalate resin A (parts by weight)	A-1 100	A-1 100	A-1 100	A-1 100	A-1 100	A-1 100
phosphazene compound B (parts by weight)	-	B-1 15	B-2 15	B-3 15	B-4 15	B-5 15
flame-retarding resin C (parts by weight)	-	-	-	-	-	-
the other flame retardant D (parts by weight)	-	-	-	-	-	-
carbonizable resin E (parts by weight)	-	-	-	-	-	-
antioxidant F (parts by weight)	F-1 0.15	F-1 0.15	F-1 F-2 0.15 0.15	F-1 F-3 0.15 0.15	F-1 F-3 0.15 0.15	F-1 F-3 0.15 0.15
drip inhibitor G (parts by weight)	G-1 0.7	G-1 0.7	G-1 0.7	G-1 0.7	G-1 0.7	G-1 0.7
filler H (parts by weight)	-	-	-	-	-	-
flame retardancy UL94	below HB	below HB	below HB	below HB	below HB	below HB

[0089]

[Table 4]

Table 4

	Comp.Ex. 7	Comp.Ex. 8	Comp.Ex. 9	Comp.Ex. 10	Comp.Ex. 11
polyalkylene terephthalate resin A (parts by weight)	A-1 100	A-1 100	A-1 100	A-2 100	A-3 100
phosphazene compound B (parts by weight)	-	-	-	-	-
flame-retarding resin C (parts by weight)	C-2 15	C-2 15	C-2 10	C-2 20	C-2 15
the other flame retardant D (parts by weight)	-	D-1 7	D-1 10	D-1 10	D-1 7
carbonizable resin E (parts by weight)	-	-	E-1 30	-	-
antioxidant F (parts by weight)	F-1 F-3 0.15 0.15	F-1 F-3 0.15 0.15	F-1 F-3 0.2 0.2	F-1 F-3 0.2 0.2	F-1 F-3 0.2 0.2
drip inhibitor G (parts by weight)	G-1 1.0	G-1 1.0	G-1 1.0	G-1 1.0	G-1 1.0
filler H (parts by weight)	-	-	-	H-1 65	-
flame retardancy UL94	below HB	below HB	below HB	below HB	below HB

[0090]

As apparent from Tables 1 to 4, polyalkylene terephthalates of Examples are remarkably improved in flame retardancy.

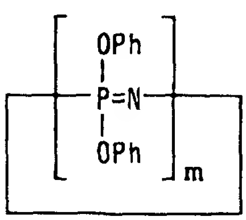
[Document Name] ABSTRACT

[Abstract]

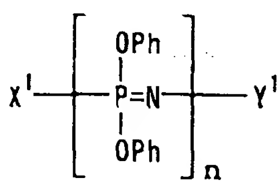
[Object(s)] To impart highly flame retardancy to a polyalkylene terephthalate resin without deteriorating the properties.

[Means to Solve the Problems] A polyalkylene terephthalate resin (e.g., polyethylene terephthalate, polybutylene terephthalate) is improved in flame retardancy by using a flame retardant containing a phenolic resin and a phosphazene compound. The phosphazene compound is a compound shown by the following formulae (1) and/or (2), and/or a compound being the above compound crosslinked with a phenylene group or a crosslinking group shown by the following formula (3). The ratio of the phosphazene compound relative to the phenolic resin (the phosphezene compound/the phenolic resin) may be about 20/80 to 80/20 (weight ratio).

[Formula 1]



(1)



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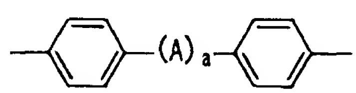

(3)

wherein X^1 represents the group $-N{=}P(OPh)_3$ or the group $-N{=}P(O)OPh$; Y^1 represents the group $-P(OPh)_4$ or the group $-P(O)(OPh)_2$; and A represents $-C(CH_3)_2-$, $-SO_2-$, $-S-$ or $-O-$.

[Selected Fig.] none

Applicant Record

Identification No.	[390006323]
1. Date of Alternation	October 22, 1990
[Reason of Alternation]	new registration
Address	3-13, Azuchi-machi 2-chome, Chuo-ku, Osaka-shi, Osaka-fu
Name	Polyplastics Co., Ltd.

Applicant Record

Identification No.	[000206901]
1. Date of Alternation	August 21, 1990
[Reason of Alternation]	new registration
Address	2-27, Ote-dori 3-chome, Chuo-ku, Osaka-shi, Osaka-fu
Name	Otsuka Chemical Co., Ltd.